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Experimental Validation for Atomistic Simulations of the Deformation of Tantalum

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Introduction and background

The transition metals exist in several crystal structures due to the influence of the d-bands on bonding. The central transition metals are stabilized in the body-centered-cubic (BCC) structure due to the approximately half filled d-bands. The d-bands have an inherent structure which imparts a directional dependence to the interatomic interactions [1]. These electronic effects, through their influence on the core structure of dislocations, cause the unusual mechanical properties observed in these metals [2].

Atomistic simulation offers a means of understanding the structure of dislocation cores and their structural changes and motion under the influence of applied stress. The interatomic distances and angles experienced by atoms residing in the core of a dislocation are highly disturbed from those in equilibrium lattice positions. A key component to reliable simulations of dislocation behavior is a proper representation of the core [3, 4]. Experimental validation of the predictions of atomistic simulations is highly desirable, yet there are few examples of successful characterization of the core structure of dislocations [5, 6]. The important dislocation in the BCC structure is the screw dislocation [2], which presents even more difficulties for characterization by high resolution transmission electron microscopy (HREM) [7]. However, grain boundaries provide an opportunity to study crystal defects possessing atoms residing in highly disturbed structures in a configuration more easily characterized by HREM and thus achieve the goal of experimental validation of atomistic simulations.

Atomistic simulations vary in complexity according to the approximations implicit in each approach. Hence, the number of atoms which can be handled in a simulation ensemble is determined by the available computational power. With modern computers, it ranges from about 10 atoms for the most sophisticated self - consistent, *ab - initio* techniques to about 10^9 atoms for simple pair - potentials. Dislocations are an example of a crystal defect with a long range strain field. The correct choice of size and boundary conditions for a simulation ensemble is critical for achieving representative behavior. The desired number of timesteps to be simulated is also a severe constraint on the size of a simulation. At least five steps per Debye period is needed, giving times for a simulation on the order of picoseconds. Hence, the modeler must perform an optimization of these parameters to achieve the desired goals. The most important choice is that of the appropriate model of the interatomic interaction to obtain representative material behavior.

The Embedded Atom Method (EAM) [8, 9] and the Finnis - Sinclair (FS) [10] method are equivalent approaches to representing the interatomic interactions and both are categorized as central force methods. They have been used with great success to predict some unexpected behaviors in face - centered - cubic metals [11, 12]. The force - matching implementation of central force potentials has even had success modeling aluminum, with its unusually high stacking fault energy [13]. Central - force potentials have also been developed for BCC metals [14], but problems with predicting correct surface reconstructions in W and Mo were quickly identified [15]. The Model Generalized Pseudopotential Theory (MGPT) [16, 17] incorporates angularly dependent interatomic interactions by including three and four body interaction terms and it is able to properly predict the (100) surface reconstruction of W [18] at the cost of needing approximately a factor of 40 times more computer time to run.

To further investigate the differences between the EAM and MGPT potentials, a comparative study was performed on the $\Sigma 5$ (310)/[001] symmetric tilt grain boundary (STGB) in niobium [19]. The two methods differed in their predictions of the relative translational state of the crystals on either side of the boundary. The MGPT potential predicted a boundary structure that possessed mirror symmetry at the atomic scale. The EAM potential predicted two low energy boundary structures in which the crystals on either side of the boundary are shifted with respect to one another by a fraction of a lattice constant, breaking the mirror symmetry of the boundary at the atomic scale. The HREM results on the $\Sigma 5$ (310)/[001] STGB in Nb showed unambiguously that mirror symmetry of the boundary was preserved [19], thus revealing another success of the MGPT versus the EAM.

A recent investigation has been reported [20] of the $\Sigma 5$ (310)/[001] and $\Sigma 5$ (210)/[001] STGBs in molybdenum. The boundaries were simulated using a FS type potential and no shifts were found for the low energy configuration for either boundary; both boundaries were predicted to be mirror symmetric at the atomic scale. The boundaries were then fabricated and their atomic structure characterized by HREM. The (210) STGB was imaged in two projections, parallel to the tilt axis and perpendicular to the tilt axis. Mirror symmetry was found for the (210) STGB. The (310) STGB was only imaged parallel to the tilt axis and again mirror symmetry was found in this projection. But no information from experiments was obtained for shifts along the tilt axis, which are not revealed by projections of the structure in that direction. Thus the experimental results for the (310) boundary were inconclusive. However, recent *ab - initio* electronic structure calculations for the (310) STGB [21] and the (210) STGB (Elsässer and Ochs, to be published) in Mo indicate a mirror symmetric structure for the (210) STGB but a shifted structure for the (310) STGB along the tilt axis.

Purpose of the project

The purpose of the project was the validation of the predictive abilities of the newly developed MGPT potentials in simulating the defect structures of BCC metals. The validation of these potentials will allow them to be applied with confidence in the simulation of materials behaviors under conditions that are not easily accessible to experimental confirmation.

Activities and approach

The approach taken and activities performed are fully detailed in the accompanying publications [22, 23] (UCRL-JC-132639 and UCRL-JC-137082, respectively). Briefly, the approach taken was to create experimentally exactly the same crystal defect as was modeled theoretically. The crystal defect chosen was the $\Sigma 5$ (310)/[001] STGB. Two different BCC metals were studied, Mo and Ta. The STGB was fabricated by UHV diffusion bonding precisely oriented single crystals. The atomic structure of these boundaries was determined by high resolution transmission electron microscopy in the new TEM facility in the CMS Directorate. The comparison of the predicted atomic structures with the experimental observations was done by simulating high resolution images using the predicted structures as initial conditions.

Technical outcome

The first publication addresses an experimental study of the $\Sigma 5$ (310)/[001] STGB in molybdenum [22]. The full three dimensional structure of the boundary was determined and a shift along the [001] direction of 1/4 a_0 was found, in agreement with MGPT simulations performed as part of the study and the electronic structure calculations of Elsässer, but in disagreement with the FS calculations of Bacia, et al.

The second paper [23] reports on the results for the atomic structure of the $\Sigma 5$ (310)/[001] STGB in Ta, determined experimentally and simulated using the MGPT. Again, agreement with the predictions of the MGPT is found.

Taken together, these results are a strong validation of the accuracy and reliability of the MGPT potentials. These potentials will now be used with confidence for the prediction of atomic structure of dislocation cores in BCC metals. The core structure of a dislocation is almost impossible to characterize experimentally [7]. But now with validated potentials the core structure can be accurately simulated, moreover it can be simulated for the more relevant condition of under an applied stress. These calculations will yield important materials parameters for the higher scale models of metals dynamics.

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References

[1] A. G. Marinopoulos, V. Vítek, and A. E. Carlsson, Significance of Non - Central Forces in Atomistic Studies of Grain Boundaries in BCC Transition Metals. *Philos. Mag. A, 72* (1995) 1311-1330.

[2] J. W. Christian, Some Surprising Features of the Plastic Deformation of Body - Centered Cubic Metals and Alloys. *Metall. Trans. A, 14A* (1983) 1237-1256.

[3] V. Vítek, R. C. Perrin, and D. K. Bowen, The Core Structure of 1/2<111> Screw Dislocations in BCC Crystals. *Philos. Mag., 21* (1970) 1049-1073.

[4] V. Vítek, Theory of the Core Structure of Dislocations in Body - Centered - Cubic Metals. *Crystal Lattice Defects*, *5* (1974) 1-34.
[5] M. J. Mills and P. Stadelmann, A Study of the Structure of Lomer and 60-Degrees Dislocations in Aluminium Using High-Resolution Transmission Electron Microscopy. *Philos. Mag. A*, *60* (1989) 355-384.
[6] N. Baluc, H. P. Karnthaler, and M. J. Mills, Tem Observation of the Fourfold Dissociation of Superlattice Dislocations and the Determination of the Fault Energies in Ni3(Al, Ta). *Philos. Mag. A*, *64* (1991) 137-150.
[7] W. Sigle, High-resolution electron microscopy and molecular dynamics study of the (a/2)[111] screw dislocation in molybdenum. *Philos. Mag. A*, *79* (1999) 1009 – 1020.
[8] M. S. Daw and M. I. Baskes, Semiempirical, Quantum Mechanical Calculation of Hydrogen Embrittlement in Metals. *Phys. Rev. Lett.*, *50* (1983) 1285-1288.
[9] M. S. Daw and M. I. Baskes, Embedded - atom Method: Derivation and Application to Impurities, Surfaces, and Other Defects in Metals. *Phys. Rev. B*, *29* (1984) 6443-6453.
[10] M. W. Finnis and J. E. Sinclair, A Simple Empirical N - body Potential for Transition Metals. *Philos. Mag. A*, *50* (1984) 45-55.
[11] F. Ernst, M. W. Finnis, D. Hofmann, T. Muschik, U. Schönberger, U. Wolf, and M. Methfessel, Theoretical Prediction and Direct Observation of the 9R Structure in Ag. *Phys. Rev. Lett.*, *69* (1992) 620-623.
[12] C. Schmidt, M. W. Finnis, F. Ernst, and V. Vítek, Theoretical and experimental investigations of structures and energies of Sigma = 3, [112] tilt grain boundaries in copper. *Philos. Mag. A*, *77* (1998) 1161-1184.
[13] F. Ercolessi and J. B. Adams, Interatomic Potentials from First - Principles Calculations: the Force - Matching Method. *Europhys. Lett.*, *26* (1994) 583-588.
[14] R. A. Johnson and D. J. Oh, Analytic Embedded Atom Method Model for BCC Metals. *J. Mater. Res.*, *4* (1989) 1195-1201.
[15] A. M. Guellil and J. B. Adams, The Application of the Analytic Embedded Atom Method to BCC Metals and Alloys. *J. Mater. Res.*, *7* (1992) 639-652.
[16] J. A. Moriarty, Analytic Representation of Multi - Ion Interatomic Potentials in Transition Metals. *Phys. Rev. B*, *42* (1990) 1609-1628.
[17] J. A. Moriarty, First - Principles Interatomic Potentials in Transition Metals: Multi - Ion Interactions and Their Analytic Representation, in R. N. Nieminen, M. J. Puska and M. J. Manninen (eds.), *Many - Atom Interactions in Solids*, Springer - Verlag, Berlin, 1990, p. 158-167.
[18] J. A. Moriarty and R. B. Phillips, First - Principles Interatomic Potentials for Transition Metal Surfaces. *Phys. Rev. Lett.*, *66* (1991) 3036-3039.
[19] G. H. Campbell, S. M. Foiles, P. Gumbsch, M. Rühle, and W. E. King, Atomic Structure of the (310) Twin in Niobium: Experimental Determination and Comparison to Theoretical Predictions. *Phys. Rev. Lett.*, *70* (1993) 449-452.
[20] M. Bacia, J. Morillo, J.-M. Pénisson, and V. Pontikis, Atomic Structure of the Σ=5, (210) and (310), [001] Tilt Axis Grain Boundaries in Mo: A Joint Study by Computer Simulation and High - Resolution Electron Microscopy. *Philos. Mag. A*, *76* (1997) 945-963.
[21] C. Elsässer, O. Beck, T. Ochs, and B. Meyer, Ab - Initio Determination of the Atomic Structure of Symmetric Tilt Grain Boundaries in BCC Transition Metals, in S. R.

Phillpot, P. D. Bristowe, D. G. Stroud and J. R. Smith (eds.), *Microscopic Simulation of Interfacial Phenomena in Solids and Liquids*, Materials Research Society, Warrendale, PA, 492, 1998, p. 121-126.

[22] G. H. Campbell, J. Belak, and J. A. Moriarty, Atomic Structure of the $\Sigma 5$ (310)/[001] Symmetric Tilt Grain Boundary in Molybdenum. *Acta Mater.*, *47* (1999) 3977 - 3985.

[23] G. H. Campbell, J. Belak, and J. A. Moriarty, Atomic Structure of the $\Sigma 5$ (310)/[001] Symmetric Tilt Grain Boundary in Tantalum. *Scripta Mater.*, (submitted to) .